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High Temperature Thermocouples for In-Pile Applications

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ABSTRACT

Traditional methods for measuring temperature in-pile degrade at temperatures above 1080 ºC. Hence, a project has been initiated to explore the use of specialized thermocouples that are composed of materials that are able to withstand higher temperature, in-pile test conditions. Results from efforts to develop, fabricate, and evaluate the performance of these specialized thermocouples are reported in this paper. Candidate materials were evaluated for their ability to withstand irradiation, to resist material interactions, and to remain ductile at high temperatures. In addition, candidate thermocouples were evaluated based on their resolution over the temperature ranges of interest. Results from these evaluations are reported, and additional on-going development activities are summarized.

KEYWORDS

High temperature measurement, thermocouples, in-pile instrumentation.

1. INTRODUCTION

To resolve principal technical and scientific obstacles to the long-term future use of nuclear energy, new reactor designs must offer enhanced safety and overcome issues involving resistance to proliferation, economics, and nuclear waste disposition. To meet these goals, new materials are being considered for fuel, cladding, and structures in advanced and existing nuclear reactors. However, there are insufficient data to characterize the performance of these new materials in high temperature, oxidizing, and radiation conditions. To evaluate candidate material performance, robust instrumentation is needed that can survive proposed test conditions. For example, traditional methods for measuring temperature in-pile degrade at temperatures above 1080 ºC. Hence, a new approach was explored which uses specialized thermocouples composed of materials that are able to withstand proposed test conditions.

This effort is the first task of a three-year project to explore and evaluate methods that are capable of measuring temperature, thermal conductivity, and deformation in-pile. In each task, promising irradiation-resistant techniques are evaluated in INL's High Temperature Test Laboratory (HTTL) to demonstrate that components can survive the temperature ranges of interest and produce data with the required accuracy. Results from initial tasks in this effort are summarized in this paper. Additional details are reported in Rempe and Wilkins (2005).

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1.1 ATR Test Conditions

This project focuses on methods that are economical, reliable, and able to obtain data for the expected/planned test conditions at INL's Advanced Test Reactor (ATR). Although specific conditions for GEN IV and AFCI fuel testing have not been finalized, ATR locations where fuels and materials are typically tested, suggest that the test conditions/requirements/geometries listed in Table 1 are of interest. In addition, typical accuracy requirements for temperature are provided.

Parameter	Specification	
	Multiple Irradiation Capsule Experiment (MICE)	Standard In-Pile Tube (SIPT)
Thermocouples		
Diameter, mm (inches)	1.63 (0.0625)	1.63 (0.0625)
Configuration	Swaged to a fixed metal sheath[a]	Swaged to a fixed metal sheath[a]
Capsule		
Dimensions		
Inner Diameter, cm (in)	2.54 (1)	2.54 (1)
Inner Height, cm (in)	20.32 (8)	20.32 (8)
Atmosphere	Helium / Neon[b]	Subcooled Water
Flowrate, gpm	low	20-60
Design flowrate, gpm	low	80
Pressure, MPa (psig)	0.10-0.17 (15-25)	12-15 (1800-2200)
Design Pressure, MPa (psig)	0.2 (30)	17 (2500)
Peak Thermal Flux ($n/cm^2/s$)[c]	4 x10^{14}[c]	4 x10^{14}
Temperature Measurement Requirements		
Range, °C	20-1300[d]	20-1300[d]
Accuracy,%	±2%	±2%
Resolution, °C	± 3	± 3
Response, msec	500	500
Rate of change, °C/sec	<< 1 (increasing)	2.4 (decreasing)

a. Required to exit through a Conax connection to maintain RCS pressure boundary.
b. Initial efforts focus on MICE irradiations in Helium and Neon. However, later efforts will consider component performance in oxidizing conditions
c. Assumed 18 MWt power level in a lobe (lobe design power level is 66 MWt); tests are assumed to last up to a year.
d. Although development efforts focus on 1300 ºC as a maximum temperature, some tests were also conducted to evaluate component performance at higher temperatures anticipated for AFCI and GEN IV fuel testing.

Table 1: Anticipated test conditions / design criteria.

1.2 Temperature Measurement Methods

Table 2 lists types of instrumentation that might be employed to measure temperature for Table 1 test conditions and design requirements. Table 2 also summarizes test temperatures, experimental error, past experience and concerns for various types of temperature instrumentation. For the proposed temperatures and radiation conditions, information in Table 2 suggests that instrumentation methods applicable to the conditions identified in Table 1 are limited to thermocouples (with thermoelements consisting of molybdenum, niobium, or zirconium, or their alloys), Johnson Noise Power Thermometers (JNPT), and ultrasonic thermometers. Although ultrasonic thermometry and JNPT techniques may be viable, it isn't clear that instrumentation capable of measuring considerably higher temperatures, such as ultrasonic thermometry, is needed. In addition, as noted in Table 2, the design of the probe and signal processing equipment for these systems is more complex (and thus, more expensive). Optical pyrometer techniques were eliminated because no viewing port is available for the proposed test conditions. Initial research suggested that optical fiber methods were not viable because progressive darkening of fibers under irradiation led to a loss of signal. Data corrections on optical fibers signals are test- and radiation-spectrum specific. Although more recent in-pile investigations suggest that optical fibers may be viable, available information suggest that more development and assessment is needed to obtain appropriate correction factors for radiation effects [Rempe and Wilkins, 2005].

Method	Temperature Range	Experience	Concerns /Comments
Chromel / Alumel "Type K"	Up to 1077-1200 °C	Successfully used for high irradiation / long term operation in inert and oxidizing media. Tolerance ~ 2-5 ºC or 0.75%	Frequent failures, drift at temperatures above ~1000 to 1100°C
Tungsten/Rhenium alloys (W-5%Re/W-26%Re) "Type C"	Up to 2200-2400 °C	Successfully used for high temperatures in inert and reducing media. Tolerance ~ 4.5°C or 0.75%	Up to 35% calibration change at neutron fluence of 2 x 10^{21} neutrons/cm^2 due to transmutation (W-Re transmutes progressively to W-Re-Os). Efforts to correct unsuccessful (corrections not applicable to other thermo-elements or test conditions).
Platinum / Rhodium "Types R, S, and B"	Up to 1760°C for Types R and S; up to 1820°C for Type B	Successfully used for high temperatures in inert or oxidizing media. Tolerance ~1.5 ºC or 0.25%	Decalibration due to transmutation of rhodium, which has a large neutron absorption cross section. Problems comparable to W/Re.
Johnson Noise Power Thermometer (JNPT)	Up to about 3000°C	Mostly experimental	Efforts to correlate the Johnson noise voltage to temperature successful for temperatures below 1500 °C, but require sophisticated and expensive signal processing.
Molybdenum/ Niobium- 1% Zirconium (Mo/Nb- 1% Zr with alumina insulators and Mo or W-22% Re sheaths)	Temperature calibration data available up to 1200ºC (but believed viable up to 1730 ºC).	Tested in the Fast Flux Test Facility MOTA at 1070 and 1375 K. Used 0.24 mm-diameter wires.	Less susceptible to irradiation. Thermocouples subjected to a total fluence of 12.5 x 10^{22} n/cm^2 and a fast fluence of 7.5 x 10^{22} n/cm^2.
Ultrasonic Thermometers	Up to 3000 °C.	Used for INEEL Power Burst Facility (PBF) test fuel centerline temperature measurements. Research following these tests investigated improvements possible with single crystal tungsten sensors.	Requires expensive and complex signal processing.
Optical Pyrometers	600 to 3000 ºC	Used for measuring surface temperatures. Error ±0.6% Small viewing section (between 0.8 to 1.5 mm)	Requires line-of-sight viewing path.
Optical Fibers	up to 800 ºC	Irradiated in Japanese Materials Test Reactor (JMTR) for thermal fluxes of 8 x 10^{16} n/m^{2}s; Irradiated in the Belgium BR1 and BR2 materials test reactor for thermal fluences up to 10^{19} n/m^2.	Data corrections required for optical fibers that are test- and radiation-spectrum specific.

Table 2: Selected high temperature measuring techniques for in-pile measurements.

The various methods for measuring temperature listed in Table 2 suggest that specialized thermocouples may be the simplest and most economic approach for in-pile high temperature measurements. Because techniques for fabricating and evaluating such thermocouples [Cannon, 1982; Knight and Greenslade, 1992; Schley and Metauer; Wilkins, 1988; and Wilkins; 1978] were last explored over 10 years ago, initial tasks for this project were devoted to developing improved versions of these earlier techniques.

2. THERMOCOUPLE DESIGN AND FABRICATION

2.1 Design

When a metallic wire is situated in a temperature gradient, a difference of electric potential exists over the temperature gradient region. The magnitude and sign of that potential difference are primarily functions of temperature and metallic composition. A thermocouple consists of two dissimilar metals selected so as to produce a single-valued thermoelectric signal of practical amplitude and suitable for use in a particular environment. Standard thermocouples, composed of different combinations of metals, have been established for industrial use to meet a range of practical considerations. Each is suitable for a particular temperature range and environmental conditions. Reliability may vary with the diameter of the wire used in the thermocouple. In practice, reliable temperature measurements are obtained from such standard thermocouples as long as the instrument's components remain unchanged. Excessive temperatures can produce progressive contamination or metallurgical changes in the thermoelements, however, that lead to calibration drift. Hostile environments, such as nuclear irradiation that transmutes the thermoelement materials, likewise produce thermoelectric decalibration and unacceptable uncertainty in the temperature measurements. Protective sheaths can attenuate some problems related to the operating environment, but they are ineffective in other cases and can even contribute to the problem of decalibration in still other situations.

As depicted in Figure 1, sheathed thermocouple probes are available with three junction types: grounded, ungrounded or insulated, and exposed. A grounded measuring junction is an integral part of the protective thermocouple sheath at its tip. It is typically produced by welding the thermoelement tips into the metal sheath closure weld. Such a configuration results in good heat transfer from the surrounding environment to the thermocouple junction. An ungrounded measuring junction is insulated from the protective thermocouple sheath at its tip. Heat transfer to the measuring junction is inhibited in this case, resulting in slower response time in comparison to a grounded junction instrument. Strain due to differential thermal expansion with the protective sheath may be increased, sometimes increasing instrument failure rates. An exposed junction protrudes from the tip of the protective thermocouple sheath and is exposed to the surrounding environment. This type offers the best response time, but is generally limited to use in non-corrosive and non-pressurized applications.

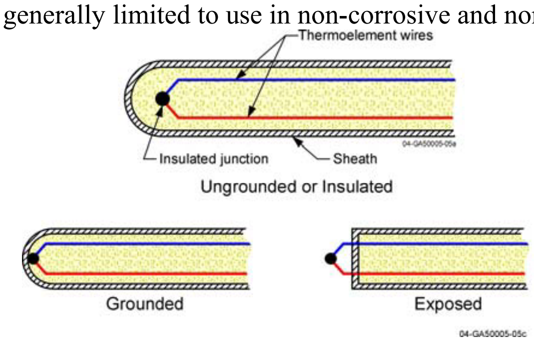


Figure 1: Thermocouple components and measuring junction configurations.

The thermoelectric output of a thermocouple must be a stable single-valued function of temperature and large enough for adequate temperature resolution. Thermocouples make use of the fact that the electromotive force (EMF) between two dissimilar metals is a function of the temperature difference (gradient) along their length. The change in EMF with respect to a change in temperature is called the Seebeck coefficient or thermoelectric sensitivity. This coefficient is usually a nonlinear function of temperature. Standard thermocouple types are manufactured to meet accepted temperature-emf curves, such as the W-Re alloy combination shown in Figure 2 (with a zero point reference junction).

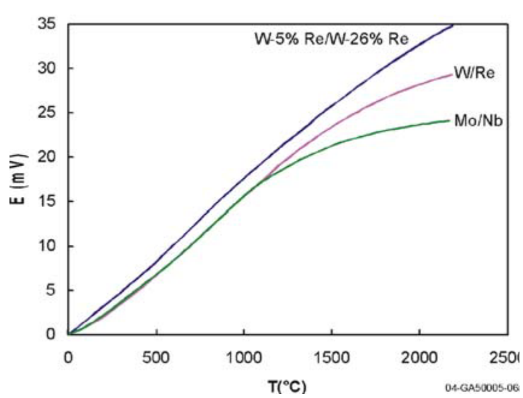


Figure 2: Typical calibration curves for Mo/Nb, W/Re and W-5% Re/ W-26% Re thermocouples.

2.2 Material Properties

As indicated above, instrumentation placed in the ATR must be able to withstand high temperatures, oxidizing atmospheres, and irradiation conditions. In addition, it is desirable to select less expensive materials for these components, when possible. Finally, materials for thermocouple components must be compatible, exhibiting similar growth and contractions with temperature. Material property information of interest for candidate thermocouple materials evaluated in this project are summarized in Tables 3 and 4.

Element	σ, barns	Melting Temperature, °C	Materials Interaction Temperature, °C								
			Mo	Nb	Pt	Re	Rh	Ta	Ti	W	Zr
Insulators[a]											
Alumina	0.46	2040	>1800	>1800	>1770	>2000	>1960	>1650	>1650	>1900	>1200
Hafnia	98.8	2810	>2200	>2200	>1770	>2200	>1960	>1872	>1650	>1700	>1800
Zirconia	0.19	2710	>1900	>1600	>1770	>1700	>1960	>1790	>1650	>1700	>1800
Metals for Thermocouples for Wires or Sheaths, or Other Components[b]											
Molybdenum	2.65	2610	NA	>2470	>1770	>2510	>1940	2610	>1670	>2470	>1550
Niobium	1.15	2470	>2470	NA	>1700	>2160	>1500	2470	>1670	>2470	>1740
Platinum	10	1770	>1770	>1700	NA	>1770	>1770	>1760	>1310	>1770	>1150
Rhenium	86	3180	>2510	>2160	>1770	NA	>1960	>2690	>1670	>2825	>1590
Rhodium	150	1960	>1940	>1500	>1770	>1960	NA	>1740	>1300	>1960	>1070
Tantalum	21.0	3017	2610	2470	>1760	>2690	>1740	NA	1668	3017	1852
Titanium	6.09	1668	>1670	>1670	>1310	>1670	>1300	1668	NA	>1670	>1540
Tungsten	18.5	3410	>2470	>2470	>1770	>2825	>1960	3017	>1670	NA	>1735
Zirconium	0.185	1852	>1550	>1740	>1150	>1590	>1070	1852	>1540	>1735	NA

a. Insulator materials data based on Economos and Kingerly (1953), Rempe, et. al. (2001), Rempe, et al. (2002), and Wilkins (1992).
b. Materials interaction data primarily based on ASM (1973). NA denotes "Not Applicable".

Table 3: Neutron capture cross sections and melting temperatures for candidate component materials

Metals	Oxidation Resistance[a]	Thermal Expansion α, 10^{-6} K-1	Electrical Resistance at 1300°C, μOhm-cm	Machinability / Weldability / Notes
Molybdenum	Susceptible to oxidation	5.1	33	dissolves readily in glass, adheres to glass, welded conventionally, machining can be difficult
Niobium	Susceptible to oxidation	7.0	62	good ductility, fair weldability, fair machinability, cost $40/kg
Platinum	Resistant to oxidation	9.0	47	rare, expensive, and ductile
Rhenium	Susceptible to oxidation	6.5	100	rare, expensive, ductile, not machinable by conventional means, weldable, usually used in alloy form, $250/oz
Rhodium	Resistant to oxidation	8.0	38	very expensive- $1000/oz
Tantalum	Susceptible to oxidation	6.5	65	galls easily; good weldability
Titanium	Susceptible to oxidation	8.5	152	ductile, good weldability, widely used, cost $50/kg
Tungsten	Susceptible to oxidation	4.4	45	machining very difficult, welding difficult, cost $25/kg
Zirconium	Susceptible to oxidation	5.8	122	good machining, good welding w/ cover gas, cost $150/kg

a. Oxidation resistance evaluated based on information in ASM International (1996), Rempe (2001), Rempe (2002), Touloukian (1967), and Wilkins (1992).

Table 4: Additional properties of interest for component thermocouple materials.

In addition to melting temperature, Table 3 lists temperatures where component materials may undergo eutectic reactions and their neutron absorption cross sections. Concerns about transmutation render materials with lower absorption cross sections more desirable. For example, tungsten and rhenium have very high melting temperatures. However, molybdenum and niobium may be preferable, not only because their melting temperatures are above the temperatures of interest (1300 ºC) but also because their lower cross sections indicate that they are less susceptible to neutron absorption and transmutation. Table 4 provides information about oxidation resistance, thermal expansion, cost, and machinability.

2.3 Approach for Evaluating Candidate Thermocouple Materials

Initial testing focused on testing sheathed thermocouples with a 0.15875 cm (0.0625 inch) outer diameter. Materials considered for each thermoelement component are listed in Table 5. Available information (see Tables 3 and 4) indicates that these materials are suitable for high temperature irradiation conditions. In addition, all of these materials have been widely used for in-pile applications. An ungrounded measuring junction was pursued in this project because it offers greater reliability and because fast response times are not required in ATR tests. In addition, a laser-welded transition splice to a stainless steel-sheathed extension cable was selected because it would permit pressure-boundary penetration via existing ATR Conax connectors.

Component	Candidate Materials
Thermoelement	Molybdenum,[a] Molybdenum-1.6% Niobium, Molybdenum (doped with Silicon and Potassium), and Molybdenum (containing Lanthanum Oxide), Zircaloy-4, Titanium-45% Niobium, Niobium-1%Zirconium
Insulator	Aluminum Oxide, Hafnium Oxide
Sheaths	Titanium, Zircaloy-4, Niobium-1%Zirconium

a. Several types of molybdenum alloys and doped molybdenums were explored to evaluated variations in the material's ductility when impurities are added.

Table 5: Candidate thermocouple materials

A typical process used to fabricate thermocouple includes:
- Clean the sheath tubing and thermoelement wires thoroughly with solvents and bakeout methods.
- Bake and outgas insulator beads.
- String insulator beads on thermoelement wires and insert into sheath tube
- Swage assembly to final diameter and anneal
- Laser-weld the measuring junction on the thermoelement tips
- Fill the sheath tip with powdered insulation to produce an insulated measuring junction
- Laser-weld a closure cap on the sheath

For initial tasks in this project, a laser-welded splice, similar to that used for zircaloy/tantalum-sheathed cladding thermocouples (Wilkins and Seopold, 1985), was pursued for joining the prototype thermocouple to a stainless steel-sheathed extension cable. Fabrication of the extension cable, with thermoelements matching the prototype thermocouple, was not a part of the initial project's workscope. However, this activity has been completed as part of another research program. Prototypes for testing in ATR to evaluate performance under irradiation was likewise not included in this initial project. However, it is anticipated that in-pile testing will be completed as part of another research program during the next year.

Thermocouple fabrication and evaluation tasks were completed at INL's High Temperature Test Laboratory (HTTL). As shown in Figure 3, the HTTL has several tube furnaces, desiccators, inert gas gettering furnaces, a sand blaster, a laser welder, and swagers available for thermocouple fabrication and testing.

Prior to prototype fabrication, materials testing was performed to provide insights for material selection. Tests were conducted, using representative thermocouple samples, to evaluate the potential for materials interactions between insulation materials and candidate sheath and thermoelement materials. To provide insight about thermoelement embrittlement and its implications for thermocouple reliability, simple mandrel-wrap tests on wires exposed to temperatures up to 1600°C were completed. Calibration tests were performed at temperatures up to 1600°C for the most promising thermoelement combinations. Results from materials interaction tests, ductility tests, and calibration tests are reported in Section 3.

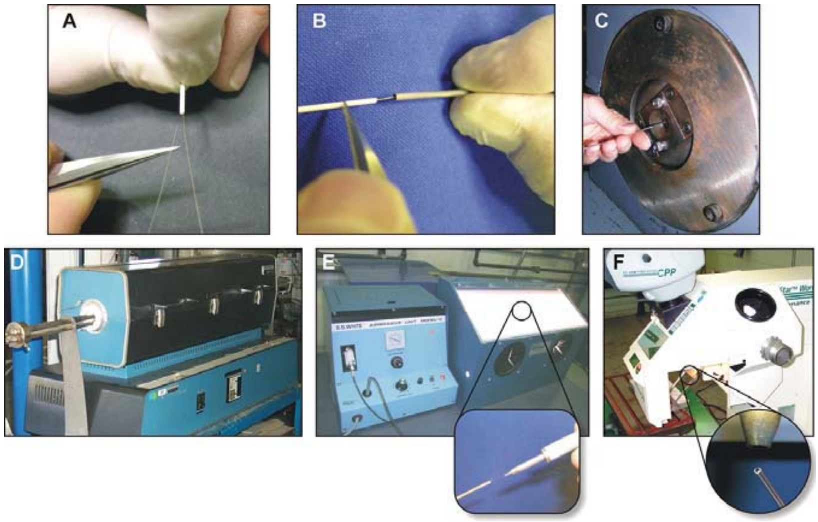


Figure 3: Thermocouple fabrication and testing activities at HTTL include: [A] insulator threading; [B] sheath loading; [C] swaging; [D] annealing; [E] sandblasting; and [F] laser welding.

3. RESULTS

3.1 Materials Interaction Tests

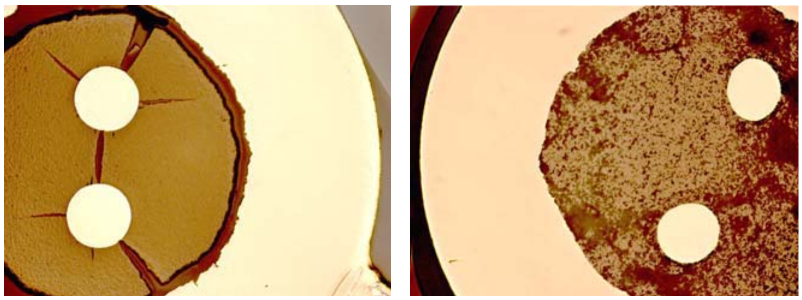
Materials interaction tests were completed by heating samples fabricated from candidate materials in gettered argon at 1300 and 1600 °C. Samples were heated for 30 minutes and then slowly cooled in an inert atmosphere to preclude oxidation. Representative thermocouple samples were constructed that allowed all candidate thermoelement, insulator, and sheath material combinations listed in Table 5 to be evaluated. As noted in Section 2, tasks were focused to ultimately develop thermocouples with an outside diameter of 0.15875 cm (0.0625 inches). After testing, samples were sectioned and Scanning Electron Microscope (SEM) analyses were performed as deemed necessary.

After testing, samples were sectioned and SEM analyses were performed as deemed necessary. As shown in Figure 4, 1300 °C test results showed that significant materials interactions occurred in samples containing Zircaloy-4 thermocouple wires and Zircaloy-4 sheaths, irrespective of the insulator material selected. SEM evaluations for one of the wires of the Zr-4 - Al_2O- Zr-4 sample indicate that the inner ring of a wire was nearly 100% zirconium. However, as one moves outward from the center of the wire, increasing concentrations of aluminum were measured (with a maximum concentration of 25 wt% near the outer surface of the wire).

(a) Zr-4 –Al_2O_3 – Zr-4 (200X) (b) Zr-4 – HfO_2 – Zr-4 (100X)

Figure 4: Results from 1300 ºC tests containing Zr-4 thermocouple wires and sheaths.

Significantly greater materials interactions were observed in many of the samples that were tested at 1600 °C. As noted in Table 3, phase diagrams suggest that significant interactions may occur between alumina and candidate thermoelement wire and sheath materials at this temperature. The gap between the alumina insulation and sheath material suggests interactions occurred in the sample shown in Figure 5(a). However, test results for Nb-1%Zr and Mo thermoelement wires and Nb-1%Zr sheaths indicate that no interactions occurred between these materials and hafnia insulators. As indicated in Figure 5(b), sheath/insulator and wire/insulator interfaces show that no discernible material interactions occurred between hafnia and the Mo thermoelement wires or hafnia and the Nb-1%Zr sheath.



(a) Mo –Al_2O_3 – Nb-1%Zr (100X) (b) Mo – HfO_2 – Nb-1%Zr (100X)

Figure 5: Results from 1600 ºC tests containing Mo thermocouple wires and Nb-1%Zr sheaths.

Table 6 summarizes insights from material compatibility tests. As shown in this table, several thermoelement wire materials (molybdenum, niobium-1% zirconium, and titanium-45% niobium) appear viable with hafnia insulation and niobium-1% zirconium sheaths. Other materials for the sheath and insulator (alumina) may also be viable if temperatures remain below 1300 °C.

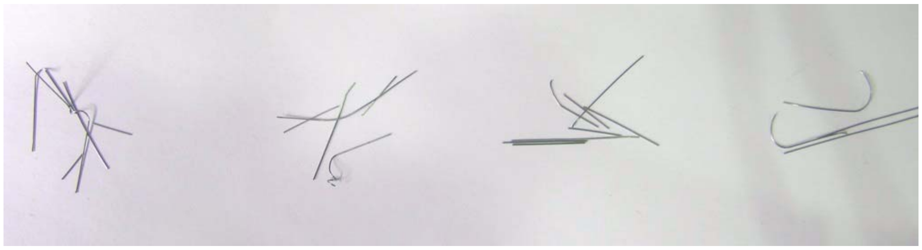
Thermoelement Wires	Insulators		Sheaths	
	1300 ºC	1600 ºC	1300 ºC	1600 ºC
Molybdenum	Hafnia, Alumina	Hafnia	Tantalum, Niobium-1% Zirconium	Tantalum, Niobium-1% Zirconium
Zircaloy-4				
Titanium-45%Niobium	Hafnia, Alumina	Hafnia	Tantalum, Niobium-1% Zirconium	Tantalum, Niobium-1% Zirconium
Niobium-1% Zirconium	Hafnia, Alumina	Hafnia	Tantalum, Niobium-1% Zirconium	Tantalum, Niobium-1% Zirconium

Table 6: Viable combinations of materials based on materials interaction tests.

3.2 Ductility Testing

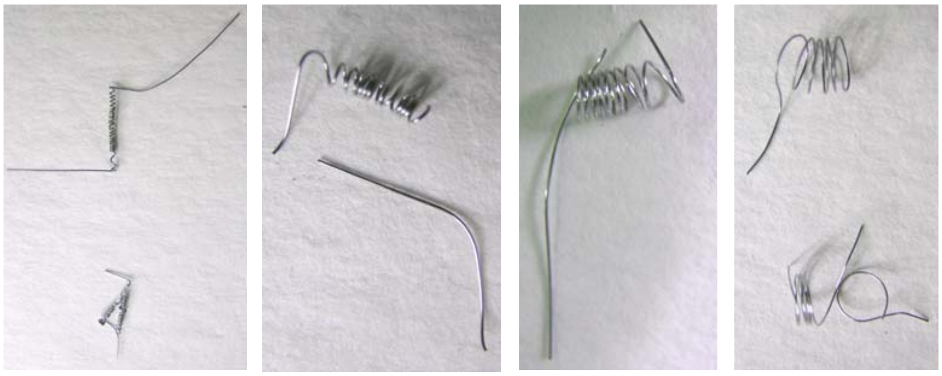
To provide insight about thermoelement wire embrittlement and its implications for thermocouple reliability, simple mandrel-wrap tests on wires exposed to temperatures up to 1600°C were completed. Table 5 lists the candidate thermoelement wires that were tested. Embrittlement samples were wrapped on mandrels of two, five, ten, and twenty times the wire diameter (e.g., mandrels with diameters of 0.0508, 0.127, 0.254, and 0.508 cm or 0.020", 0.050", 0.100", and 0.200"). Those metals that wrap without damage on a small-diameter mandrel after high-temperature exposure are better candidates from the standpoint of embrittlement.

In general, suitable ductility was observed with all of the thermocouple wires evaluated. The one exception occurred with "undoped" molybdenum wire, which will recrystallize at 1200 °C. As illustrated in Figure 6, this wire was very brittle after heating at 1300 °C. However, suitable ductility was observed in the other molybdenum wires tested (e.g., Mo doped with silicon and potassium, Mo containing lanthanum oxide, and the Mo-1.6 Nb alloy). As shown in Figure 7, these wires remained ductile even after heating at 1600 ºC for 30 minutes.



(a) 0.0.051 cm (b) 0.127 cm (c) 0.254 cm (d) 0.508 cm

Figure 6: Results from undoped Mo (0.0254 cm diameter) wire after heating at 1300 °C.



(a) 0.051 cm (top unheated) (b) 0.127 cm (c) 0.254 cm (d) 0.508 cm (top unheated)

Figure 7: Molybdenum wire doped with silicon, tungsten, and potassium (0.0254 cm diameter) after heating in argon at 1600 ºC for 30 minutes.

3.2 Thermoelectric Calibration

As discussed above, the thermoelectric output of a thermocouple must be a stable single-valued function of temperature, large enough for adequate temperature resolution, and repeatable. To further evaluate the viability of proposed candidate thermocouple materials, calibration tests were completed for each of the candidate thermocouple element combinations listed in Table 7. These combinations were selected because these materials did not undergo significant materials interactions with candidate insulator materials and were sufficiently ductile after heating (although some combinations experienced significant interactions at temperatures above 1300ºC). It should be noted that these tests are initial calibration tests to evaluate the viability of proposed thermoelement materials. Once the design of the proposed thermocouple is finalized for a particular application, additional calibration tests will be conducted that correspond to the actual thermocouple geometry and test conditions.

Positive Thermocouple Wire	Negative Thermocouple Wire
Molybdenum doped with Potassium, Tungsten, and Silicon	Niobium-1% Zirconium
Molybdenum doped with Potassium, Tungsten, and Silicon	Titanium-45%Niobium
Molybdenum doped with Potassium, Tungsten, and Silicon	Zircaloy-4
Molybdenum -1.6% Niobium	Niobium-1% Zirconium
Molybdenum -1.6% Niobium	Titanium-45%Niobium
Molybdenum -1.6% Niobium	Zircaloy-4

Table 7: Combinations of thermocouple wires for calibration tests.

Calibration tests were completed using the setup shown in Figure 8. As shown in the figure, thermoelement combinations were heated in gettered argon in a tube furnace. Data were obtained from room temperature (approximately 20 ºC) up to 1600 ºC. An ice point cell was used to obtain a 0º C reference temperature. The emf response of the candidate thermoelement combination was obtained at selected temperatures. In addition, a Type C thermocouple (W-5%Re versus W-26%Re) was used for reference temperature measurements. Note that several runs were completed to demonstrate repeatability of the thermoelectric response.

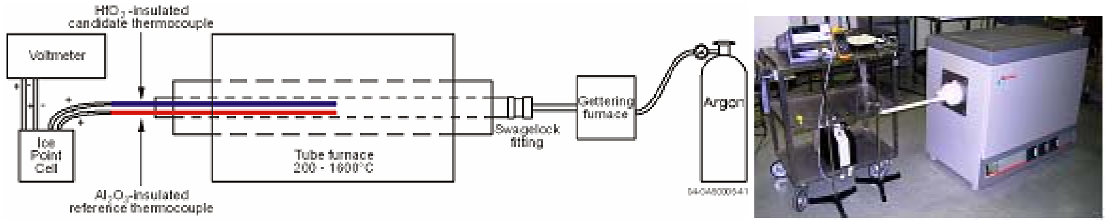


Figure 8: Calibration test setup.

Calibration results (see Figure 9) indicate that the thermoelectric response is single-valued and repeatable for all of the candidate thermoelements considered. In addition, results indicate that the high temperature resolution is acceptable for all thermocouple element combinations considered (although some combinations are limited due to materials interactions at temperatures below 1600 ºC). The selection of the thermocouple element combination will depend on the desired peak temperature and accuracy requirements. If thermocouples are needed that measure temperatures at 1600 ºC or higher, the doped Mo / Nb-1% Zirconium or Mo-1.6% Nb / Nb-1% Zirconium combination is recommended. However, it should be recognized that the above results are preliminary. Additional testing is needed to evaluate the effects of heat treating and thermal cycling.

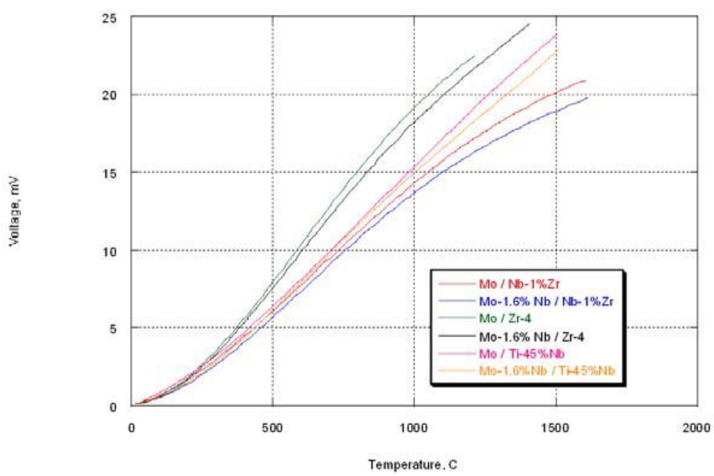


Figure 9: Comparison of calibration curves for tested thermoelement combinations.

4. CONCLUSIONS AND RECOMMENDATION FOR FURTHER EVALUATION

As discussed in this paper, a project has been initiated that explores the use of specialized thermocouples that are composed of materials able to withstand high temperature in-pile applications. Results from efforts to develop, fabricate, and evaluate the performance of these specialized thermocouples suggest that several material combinations are viable. Tests show that several low neutron cross-section candidate materials are resistant to material interactions and remain ductile at high temperatures. In addition, results indicate that the thermoelectric response is single-valued and repeatable for all of the candidate thermoelements considered. Tests indicate that the high temperature resolution is acceptable for all thermocouple element combinations considered (although some combinations are limited due to materials interactions at temperatures below 1600 ºC). The selection of the thermocouple element combination will depend on the desired peak temperature and accuracy requirements. However, if thermocouples are needed that measure temperatures at 1600 ºC or higher, the doped Mo / Nb-1%Zirconium and Mo-1.6% Nb / Nb-1%Zirconium combinations are recommended.

It is recognized that additional development activities are needed for proposed thermocouple designs. For example, stability and thermal cycling tests will help to characterize the reliability of candidate thermocouples. Because past experience has indicated calibration differences among various lots of wire, comparison tests should be performed on wire procured from several manufacturers. In addition, several application-specific tasks are also needed. For example, once the design of the proposed thermocouple is finalized for a particular application, additional calibration tests must be conducted that correspond to the actual thermocouple geometry and test conditions. In addition, activities are needed to develop a laser-welded splice for joining the prototype thermocouple to a stainless steel-sheathed extension cable. Several of these additional activities are already underway at the HTTL as part of other research programs.

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REFERENCES AND CITATIONS

American Society for Metals (ASM), 1973. "Metals Handbook," Vol. 8, *Metallography, Structures, and Phase Diagrams*, 8th Edition, American Society for Metals, Metals Park, OH, 1973.

American Society for Metals (ASM) International, 1996. *Binary Alloy Phase Diagrams*, Second Edition, Plus Updates, Version 1.0, 1996.

Cannon, C. P., 1982. "2200°C Thermocouples for Nuclear Reactor Fuel Centerline Temperature Measurements," *Temperature: Its Measurement and Control in Science and Industry,* J. F. Schooley, Ed.-in- Chief (American Institute of Physics, New York, 1982) Vol. 5, Part 2, pp 1061-1067.

Economos, G., and Kingery, W.D., "Metal-Ceramic Interactions: II, Metal-Oxide Interfacial Reactions at Elevated Temperatures," *Journal of the American Ceramic Society*, **36**, No. 12, December 1953 pp. 404-409.

Knight, R.C., and D. L. Greenslade, D. L., 1992. *Irradiation Testing of a Niobium-Molybdenum Developmental Thermocouple,* Westinghouse Hanford Company Technical Report WHC-SA-1256-FP, October 1991 (also presented at the Ninth Symposium on Space Nuclear Power Systems, 13-16 January 1992, Albuquerque, NM).

Rempe, J. L., and Wilkins, S. C., 2005. *Specialized Thermocouples for High Temperature In-Pile Applications*, Idaho National Engineering and Environmental Technical Report INEEL/EXT-05-02576, January 2005.

Rempe, J. L., et al., 2002. *NUPEC Corium Coolability Project Support - Direct Electrical Heating Investigations,* Idaho National Engineering and Environmental Technical Report INEEL/EXT-02-00030, March 2002.

Rempe, J. L., et al., 2001. *NUPEC Corium Coolability Project Support - Heater Development and Testing,* Idaho National Engineering and Environmental Technical Report INEEL/EXT-01-00372, March 2001.

Schley, R., and Metauer, G., "Thermocouples for Measurements under Conditions of High Temperature and Nuclear Radiation," T*emperature: Its Measurement and Control in Science and Industry,* J. F. Schooley, Ed.-in-Chief (American Institute of Physics, New York, 1982, **5**, Part 2, pp. 1109- 1113.

Touloukian, Y. S., Editor, 1967. *Thermophysical Properties of High Temperature Solid Materials, Volume 4: Oxides and their Solutions and Mixtures - Part I: Simple Oxygen Compounds and their Mixtures*, The MacMillan Company: New York, NY.

Wilkins, S. C., 1978. "Experimental Coaxial Molybdenum-Zircaloy Fuel Rod Cladding Surface Thermocouple," TFBP-TR-284, September 1978.

Wilkins, S. C. and Seopold, L.K., 1985, "Fabrication and Use of Zircaloy/Tantalum-Sheathed Cladding Thermocouples and Molybdenum/Rhenium-Sheathed Fuel Centerline Thermocouples," *Idaho National Engineering Laboratory Informal Report,* EGG-TFBP-6739, January 1985.

Wilkins, S.C., et al., 1988. "Low Cross-Section Mo-Nb Thermocouples for Nuclear Applications: The State-of-the-Art," *Fifth Symposium on Space Nuclear Power Systems*, pp 499-502, Albuquerque, New Mexico, January 11-14, 1988.

Wilkins, S. C., 1992. "Characterization and Materials-Compatibility Tests of Molybdenum-Niobium Thermocouples," *Seventh International Symposium on Temperature: Its Measurement and Control in Science and Industry,* Toronto, Ontario, Canada, April 28-May 1, 1992.